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**FACTORS AFFECTING SOLUBILIZATION OF  
PLUTONIUM AND AMERICIUM FROM POND  
SEDIMENTS AT THE ROCKY-FLATS FACILITY.**

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## ABSTRACT

Plutonium and americium are solubilized from sediment into pond water in increasing amounts at pH values of nine and above. They are not dissolved but rather dispersed as a colloid, whose average size decreases with increasing pH. Only a fraction of the plutonium and americium can be dispersed; moreover the pond water has a low saturation value and will gradually redeposit these elements onto the sediment. These factors suggest that migration of plutonium and americium from the ponds would be slow, and also lead to the conclusion that these elements cannot be removed efficiently from the sediment by leaching at high pH.

## INTRODUCTION

The behavior of transuranium elements - especially plutonium - in aquatic environments, both marine and fresh has received considerable attention<sup>1-4</sup>, because of nuclear weapons tests in the Pacific, and as a result of liquid emissions from nuclear energy facilities and waste-burial sites. With the exception of factors dependent on ionic strength, the behavior of plutonium and americium is similar in both sea and fresh water. In particular, both elements tend to concentrate in pond and ocean sediments<sup>5-10</sup>, but in marine systems are not permanently immobilized and under certain conditions may be resolubilized\* into the water phase<sup>8,9</sup>. The solubilities of the most common compounds of plutonium in the environment - the oxide and the hydroxide - are extremely low<sup>11</sup>; and even after due account is taken of Davydov's<sup>12</sup> excellent point that the solubility product of plutonium hydroxide is not an accurate expression of the concentration of plutonium in water, it is still apparent that this concentration can be many orders of magnitude higher than predicted by solubility considerations done.

Hydrolyzed plutonium is known to polymerize to form colloidal species<sup>11,13,14</sup>, and these are the predominant forms of the element in natural waters<sup>13</sup>. Depending on conditions, the colloidal plutonium (and americium) may (1) remain dispersed indefinitely; (2) be adsorbed

\* In this paper, the term solubilization will be used to refer to any dispersal of a solid in water, whether the mechanism be true dissolution, colloid peptization, or formation of a fine suspension.

by bottom sediment; (3) be redispersed, still in colloid form, by the water medium. Conditions which affect the behavior of plutonium and americium include pH, ionic strength, temperature, and complexing anion concentration. In fresh water systems of low salt content and ambient temperature, pH would be expected to be the most important variable.

In this paper we report the results of investigation of the behavior of plutonium and americium in a fresh water holding pond at the Rocky Flats Plant of the Energy Research and Development Administration. This pond, known as B-1, was selected for study because it was known to have appreciable concentrations of plutonium and americium in the sediment. This pond has been receiving treated sewage effluent since 1952 and laundry waste waters from this date until late 1973. Most of these elements now present in the sediment are from the latter stream, which was contaminated in the laundering of protective clothing used in a plutonium processing facility. Pond B-1 has a maximum depth of approximately four meters and a volume of  $3 \times 10^6$  liters; the residence time for water in it is approximately 34 hours\*. Although the results reported here are specific to Pond B-1, they should be at least qualitatively applicable to other fresh water ponds of similar pH and sediment composition.

\* Thompson<sup>7</sup> has described the holding pond system at Rocky Flats.

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## EXPERIMENTAL

The sediments used in this study were collected from the bottom of Pond B-1 using a coring device which penetrated only the top few centimeters of sediment. Plant fragments and other debris were removed prior to homogenizing with an electric mixer. The homogenized sediment had an average concentration of 1620 pCi Pu/g and 460 pCi Am/g based on dry weight.

The aqueous phase used in the experiments was water collected from Pond B-1 at the same time as the sediment. The water had a pH of 7.8 and contained 2.3 pCi/l plutonium and 1.6 pCi/l americium. The following anions were present in concentrations expressed as millimoles/liter: nitrate, 0.4; sulfate 1.2; bicarbonate, 0.6; total phosphate (as  $\text{PO}_4^{3-}$ ), 0.02; chloride, 0.5; fluoride, 0.03.

Size fractionation of the sediment was accomplished using a liquid sedimentation procedure based on Stokes' law:

$$U_m = \frac{2}{9} \frac{s - s'}{\mu} \cdot r^2 g$$

- where  $U_m$  = terminal settling velocity  
 $s$  = particle density (assumed to be 2.5 g/cm<sup>3</sup>)  
 $s'$  = liquid density (1.0 g/cm<sup>3</sup> for water)  
 $\mu$  = liquid viscosity (0.01 g/sec. cm. for water)  
 $r$  = particle radius  
 $g$  = gravitational acceleration (980 cm/sec<sup>2</sup>).

Extraction\* experiments were conducted at room temperature by shaking 35 ml of Pond B-1 water with B-1 sediment in 50 ml polycarbonate centrifuge tubes on a Burrell wrist-action shaker. (Before shaking commenced, the sediment-water mixtures were adjusted to the specified pH with dilute HCl or NaOH.) Generally 5 ml of sediment (equivalent to approximately 1.2 grams dry) was used in each extraction except in the runs involving filtration; in these runs, where it was desirable to minimize the amount of material retained on the filters, only one milliliter of sediment was used. At the end of the specified extraction time the tubes were centrifuged at 5000 rpm for five minutes and a five milliliter sample of the supernate withdrawn for analysis. In those experiments involving extraction for successive time intervals, the tubes were then shaken vigorously to resuspend the sediment and replaced on the shaker for additional extraction; the above procedure was then repeated at each specified time period. At the conclusion of the experiments, the residues were dried and analyzed for plutonium and americium. From these values and the concentrations of these two elements in the extraction supernates, the extraction coefficients\* ( $K_{ex}$ ) were calculated by the expression

$$K_{ex} = \frac{pCiPu(Am)/ml\ soln}{pCiPu(Am)/g\ residue}$$

In the runs in which filtration was employed the tubes were not centrifuged, but were allowed to stand for 57 minutes, the calculated time to allow particles larger than 5  $\mu$ m to settle.

\* The terms extraction and extraction coefficient are used in this paper to describe the distribution of plutonium and americium between sediment and water; these terms should not be interpreted in the strict chemical sense, however, since the plutonium and americium are not in true solution.

Nucleopore filters of the specified pore size were used in all filtrations except the  $0.4\ \mu\text{m}$  separations at pH 12, which were accomplished with  $0.45\ \mu\text{m}$  Nalge filters.

Supernate samples were evaporated on stainless steel planchets and analyzed by alpha spectrometry using the 5.15 Mev alpha peak for plutonium-239 and the 5.5 Mev peak for americium-241. The samples were counted on a Nuclear Data 4420 pulse height analysis system with ORTEC surface barrier detectors; the counting accuracy was  $\pm 6\%$  at the 95% confidence level.

The residues from the extraction runs were leached with 1:1  $\text{HNO}_3\text{-HCl}$  solution containing a small amount of  $\text{HF}$ , and plutonium was separated from americium by anion exchange. The americium fraction was evaporated on a stainless steel planchet and counted as above. The plutonium fraction was extracted with 0.5M thenoyltrifluoroacetone (TTA) in xylene, and the organic extract was evaporated and counted as described above.

Anion analyses of the pond water involved the following procedures: nitrate by a colorimetric method using phenoldisulfonic acid; sulfate by titration with  $\text{BaCl}_2$ ; bicarbonate by titration with  $\text{H}_2\text{SO}_4$ ; phosphate by the colorimetric phosphomolybdate method; and chloride and fluoride by specific ion electrodes.

## RESULTS AND DISCUSSION

The size and activity distribution of Pond B-1 sediment is shown in Table 1. These results indicate a slightly higher concentration of plutonium in particles in the 4.6 to 9  $\mu$  size range and a more uniform distribution of americium among the various sizes. To characterize the sediment further, a sample was centrifuged and the residue frozen. The frozen sediment was cut along the vertical axis and contacted directly along the cut (inner) surface with a strip of polycarbonate film for four weeks to allow alpha tracks to form in the film. Examination of the film indicated a tendency for the plutonium to concentrate near the top of the sediment, suggesting that it may be preferentially attached to the less dense portion of the sediment. Of possible relevance is the finding of Edgington<sup>10</sup> et al that most of the plutonium in Lake Michigan sediment is associated with hydrous metal hydroxides and only a small fraction with natural humates.

Concentrations of plutonium and americium in pond water after extraction with sediment for various time periods in the pH 3 to 12 range are shown in Tables 2 and 3. Each value is the mean of five replicates. Extraction remains relatively constant in the pH 3 to 8 range, but from pH 9 to 12 increases rapidly, yielding especially high concentrations of plutonium after 34 and 68 hours at pH 11 and 12. The increased solubilization at high pH may be partly a result of the change in sign of the plutonium colloid from positive to negative at pH 7.5 to 8<sup>16</sup>, if this is true it suggests that the plutonium colloid is adsorbed by negative sites and then rerelled as the pH is increased. The values at pH 9 and 10, although not as high, are perhaps of more practical concern since they are still considerably above normal and



TABLE 1. Activity and Particle Size Distribution of Pond B-1 Sediment

<u>Size Fraction, <math>\mu\text{m}</math></u>	<u>% of Total Wt.</u>	<u>Pu, pCi/g</u>	<u>% of Total Pu</u>	<u>Am, pCi/g</u>	<u>% of Total Am</u>
+ 74	0.33	382	0.08	92	0.09
+52-74	0.32	2340	0.45	224	0.20
+37-52	0.48	321	0.09	98	0.13
+26-37	2.48	573	0.86	250	1.76
+19-26	2.69	887	1.45	265	2.03
+13-19	7.42	778	3.52	294	6.21
+ 9-13	12.84	1032	8.08	301	11.00
+6.5-9	39.48	2040	49.15	355	39.90
+4.6-6.5	19.47	2105	25.00	470	26.04
+2.3-4.6	6.96	1576	6.69	329	6.52
-2.3	7.55	1000	4.61	285	6.12

are more likely to be attained by an accidental pH excursion. These results are in apparent contradiction to those of Thompson<sup>7</sup>, who found plutonium and americium to be tightly bound to the sediment; however, no pH values are stated in his paper, and it is possible that his data were obtained at pH 7 to 8 where there is minimal solubilization of these elements. The extent to which these data can be extrapolated to natural field conditions is problematical since the optimum contact between water and sediment would not be achieved in the field. Nevertheless they do indicate a potential source of water contamination that bears close scrutiny.

The values in Tables 2 and 3, although of practical significance, do not relate directly to the concentrations of plutonium and americium in the sediment. Of more fundamental interest are the results in Tables 4 and 5, in which the sediment concentrations are used to calculate extraction coefficients. The same trends are noted, and a comparison of the data in these four tables indicates that while the extraction coefficients are low, they can result in sufficient plutonium and americium solubilization to be a matter of potential concern.

The enhanced solubilization at high pH values, at which both plutonium and americium would be expected to be less soluble, suggests that they are not in true solution but rather exist as a colloidal sol whose formation by peptization of the sediment is enhanced at high pH values. To assess this possibility, a series of 20-hour extractions was conducted with four replicate samples each at pH values of 3, 8, 9, and 12. After extraction and settling, portions of the supernates were filtered through Nuclepore filters of pore sizes 5, 1, 0.4, and 0.2  $\mu\text{m}$ .

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TABLE 2. Plutonium Sediment Extraction Results

Plutonium Concentration in Supernate, pCi/l, After Extraction Time Shown

<u>Initial pH</u>	<u>2 Hours</u>	<u>4 Hours</u>	<u>8 Hours</u>	<u>20 Hours</u>	<u>34 Hours</u>	<u>68 Hours</u>	<u>Final pH</u>
3	10	22	33	29	34	55	3.4
4	20	7	9	5	26	43	3.8
5	5	8	8	3	8	34	5.1
6*	6	5	11	11	11	18	7.0
7	11	20	8	12	14	12	7.2
8	49	29	23	18	19	20	7.1
9*	123	216	179	115	36	27	7.6
10	195	310	338	669	384	36	8.6
11	220	261	354	425	1402	1202	10.7
12	105	106	205	629	913	4474	12.0

\* Values in these rows are means of four replicates; all others are means of five replicates.

TABLE 3. Americium Sediment Extraction Results

Americium Concentration in Supernate, pCi/l, After Extraction Time Shown

Initial pH	2 Hours	4 Hours	8 Hours	20 Hours	34 Hours	68 Hours	Final pH
3	13	13	18	4	18	34	3.4
4	7	5	4	2	13	19	3.8
5	4	5	4	2	5	18	5.1
6*	<1	4	4	7	7	8	7.0
7	3	6	4	4	8	10	7.2
8	19	15	7	8	10	8	7.1
9*	51	72	81	47	19	10	7.6
10	86	121	121	291	187	11	8.6
11	95	121	162	182	293	311	10.7
12	69	44	129	248	324	693	12.0

\* Values in these rows are means of four replicates; all others are means of five replicates.

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TABLE 4. Plutonium Sediment Extraction Coefficients

Plutonium Extraction Coefficient  $\times 10^6$ , After Extraction Time Shown

Initial pH	2 Hours	4 Hours	8 Hours	20 Hours	34 Hours	68 Hours
3	8.1( $\pm 0.8$ )*	13.8( $\pm 3.0$ )	21.0( $\pm 2.9$ )	18.4( $\pm 4.8$ )	22.1( $\pm 4.9$ )	34.8( $\pm 7.4$ )
4	11.8( $\pm 5.0$ )	4.3( $\pm 1.2$ )	6.1( $\pm 3.7$ )	2.8( $\pm 1.2$ )	19.5( $\pm 7.8$ )	26.8( $\pm 4.4$ )
5	3.2( $\pm 1.3$ )	8.2( $\pm 7.3$ )	5.4( $\pm 2.8$ )	2.0( $\pm 0.2$ )	5.1( $\pm 0.7$ )	22.1( $\pm 2.4$ )
6	4.3( $\pm 4.6$ )	4.1( $\pm 4.7$ )	9.7( $\pm 0.9$ )	6.9( $\pm 0.8$ )	6.2( $\pm 0.6$ )	9.2( $\pm 2.5$ )
7	8.6( $\pm 2.6$ )	13.1( $\pm 3.6$ )	5.2( $\pm 3.3$ )	6.2( $\pm 5.1$ )	9.5( $\pm 1.2$ )	8.0( $\pm 0.7$ )
8	27.7( $\pm 12.1$ )	16.8( $\pm 6.6$ )	12.1( $\pm 2.7$ )	10.0( $\pm 3.0$ )	13.6( $\pm 1.6$ )	10.9( $\pm 4.0$ )
9	86.6( $\pm 31.5$ )	149.1( $\pm 28.5$ )	124.9( $\pm 25.4$ )	81.4( $\pm 10.5$ )	25.2( $\pm 3.3$ )	19.1( $\pm 6.3$ )
10	144.2( $\pm 58.0$ )	198.2( $\pm 41.9$ )	216.6( $\pm 47.0$ )	426.6( $\pm 48.4$ )	246.1( $\pm 48.5$ )	23.6( $\pm 5.5$ )
11	132.7( $\pm 17.7$ )	179.1( $\pm 57.6$ )	226.4( $\pm 60.4$ )	245.3( $\pm 24.4$ )	848.4( $\pm 170.7$ )	656.9( $\pm 34.2$ )
12	65.1( $\pm 18.0$ )	64.5( $\pm 10.1$ )	125.6( $\pm 30.7$ )	434.8( $\pm 94.6$ )	556.5( $\pm 72.9$ )	2762.9( $\pm 699.2$ )

\* In this and all succeeding tables, values in parenthesis are standard deviations.

TABLE 5. Americium Sediment Extraction Coefficients

Americium Extraction Coefficient  $\times 10^6$ , After Extraction Time Shown

<u>Initial pH</u>	<u>2 Hours</u>	<u>4 Hours</u>	<u>8 Hours</u>	<u>20 Hours</u>	<u>34 Hours</u>	<u>68 Hours</u>
3	24.2( $\pm 6.4$ )	23.9( $\pm 5.5$ )	32.1( $\pm 4.2$ )	7.3( $\pm 6.2$ )	33.6( $\pm 9.9$ )	61.6( $\pm 17.5$ )
4	12.1( $\pm 5.1$ )	8.1( $\pm 3.7$ )	7.4( $\pm 2.2$ )	3.3( $\pm 5.1$ )	27.4( $\pm 11.8$ )	33.4( $\pm 7.4$ )
5	6.5( $\pm 6.1$ )	9.4( $\pm 1.7$ )	6.5( $\pm 5.0$ )	2.9( $\pm 2.5$ )	9.0( $\pm 3.4$ )	33.0( $\pm 5.9$ )
6	<1.0	7.4( $\pm 6.8$ )	7.4( $\pm 6.0$ )	12.7( $\pm 1.7$ )	13.5( $\pm 1.4$ )	14.6( $\pm 7.2$ )
7	5.5( $\pm 3.3$ )	15.9( $\pm 11.2$ )	5.0( $\pm 1.6$ )	6.6( $\pm 5.8$ )	14.4( $\pm 4.9$ )	18.7( $\pm 2.2$ )
8	48.4( $\pm 5.4$ )	38.9( $\pm 15.5$ )	16.9( $\pm 8.7$ )	20.5( $\pm 7.0$ )	25.9( $\pm 8.6$ )	21.9( $\pm 2.2$ )
9	135.9( $\pm 59.5$ )	192.4( $\pm 53.5$ )	216.2( $\pm 46.9$ )	104.0( $\pm 11.4$ )	51.7( $\pm 7.3$ )	25.5( $\pm 7.1$ )
10	223.9( $\pm 13.3$ )	315.2( $\pm 44.2$ )	314.6( $\pm 40.7$ )	679.6( $\pm 78.1$ )	486.6( $\pm 25.1$ )	27.8( $\pm 9.1$ )
11	272.2( $\pm 40.7$ )	351.2( $\pm 64.3$ )	468.3( $\pm 88.0$ )	522.7( $\pm 79.4$ )	840.5( $\pm 131.4$ )	892.4( $\pm 123.2$ )
12	204.6( $\pm 72.6$ )	129.4( $\pm 11.1$ )	373.4( $\pm 112.7$ )	720.6( $\pm 49.6$ )	996.7( $\pm 51.3$ )	2003.9( $\pm 518.8$ )

Results, shown in Table 6, clearly indicate that plutonium and americium are present almost entirely in colloidal form at all pH values studied. Only that concentration of these elements filterable through 0.2  $\mu\text{m}$  filters can be present in true solution, and a large portion of this fraction is undoubtedly present as smaller colloids. The plutonium concentrations in the 0.2  $\mu\text{m}$  filtrates at pH 3, 8, and 9 do not differ significantly from that of the original pond water, indicating that there was no increase in dissolution in this range due to enhanced complex formation with anions in the solution. Hence it can be concluded that degree of complexation was not a variable in this study. Americium concentrations in the 0.2  $\mu\text{m}$  filtrates are higher than in the original pond water, but this is almost certainly the result of the greater solubility of americium and a smaller colloid size rather than to complexation. It is noteworthy that the average size of the colloids decreases with increasing pH. A finding which is a direct contradiction to the results of Rhodes<sup>15</sup> who found the particle size of the colloid increased with increasing pH. Using the 5  $\mu\text{m}$  filtrate concentration as a basis of comparison, the results in Table 7 were calculated. The colloidal size distributions of plutonium at pH 3 and 8 are similar, but there is a shift to smaller size ranges at pH 9, and this shift is enhanced at pH 12. Only at pH 12 is there a significant plutonium concentration in the 0.2  $\mu\text{m}$  filtrate. In the case of americium, the trend is similar, but less regular, probably reflecting both the decreased tendency toward polymerization and the greater solubility of monomeric americium hydroxide compared to the plutonium analogue.

The relatively large solubilization of plutonium and americium at high pH values raises the question of whether the amount of these elements suspended is limited by their availability in the sediment or whether saturation of the water phase becomes the limiting factor.

TABLE 6. Filtrate Particle Size Determination

Concentration in Filtrate After Filtration Through Pore Size Shown

pH	Pu, pCi/l				Am, pCi/l			
	5 $\mu$ m	1 $\mu$ m	0.4 $\mu$ m	0.2 $\mu$ m	5 $\mu$ m	1 $\mu$ m	0.4 $\mu$ m	0.2 $\mu$ m
3	50( $\pm$ 2)	6.6( $\pm$ 2.5)	4.1( $\pm$ 1.1)	2.4( $\pm$ 0.8)	52( $\pm$ 7)	31( $\pm$ 4)	12( $\pm$ 2)	9( $\pm$ 3)
8	59( $\pm$ 6)	4.3( $\pm$ 0.9)	4.3( $\pm$ 1.7)	2.7( $\pm$ 1.0)	58( $\pm$ 10)	15( $\pm$ 3)	13( $\pm$ 2)	14( $\pm$ 2)
9	44( $\pm$ 7)	41.0( $\pm$ 6)	25.0( $\pm$ 6)	2.7( $\pm$ 1.0)	39( $\pm$ 4)	34( $\pm$ 7)	23( $\pm$ 6)	7.3( $\pm$ 3.0)
12	50( $\pm$ 2)	38.0( $\pm$ 6)	41.0( $\pm$ 6)*	8.2( $\pm$ 3.2)	39( $\pm$ 2)	23( $\pm$ 4)	5.4( $\pm$ 1.8)*	8.1( $\pm$ 5.5)

\* These samples filtered through 0.45  $\mu$ m filters.



**TABLE 7. Colloidal Size Distribution of Plutonium and Americium**

<u>pH</u>	<b>Percent* of Plutonium and Americium in Size Fraction</b>					
	<u>&lt;1 <math>\mu</math>m</u>		<u>&lt;0.4 <math>\mu</math>m</u>		<u>&lt;0.2 <math>\mu</math>m</u>	
	<u>Pu</u>	<u>Am</u>	<u>Pu</u>	<u>Am</u>	<u>Pu</u>	<u>Am</u>
3	13	60	8	23	5	17
8	7	26	7	22	5	24
9	93	87	57	59	6	19
12	76	59	82	14	16	21

\* Relative to the concentrations of plutonium and americium filterable through a 5  $\mu$ m filter.

To explore this question, four replicate samples at pH 9 and four at pH 11 were extracted for 20 hours, centrifuged, and the water removed by decantation and sampled. The sediment was again extracted for 20 hours with fresh pond water at the original pH, while the original water phase was contacted for 20 hours with fresh sediment, again at the original pH. After centrifugation and liquid phase sampling, the above procedure of contacting the original water with fresh sediment and the original sediment with fresh water was repeated two more times. The results are shown in Tables 8 and 9.

The data in Table 8 indicate that the amount of plutonium suspended decreases with each successive contacting, suggesting that only a fraction of these elements present in the sediment is available for solubilization. Perhaps this fraction is less tightly bound to the sediment, or is associated with smaller, less dense particles in the sediment.

The dispersions created by these extractions not only have a saturation value, but are unstable, as indicated by Table 9. The decrease in plutonium and americium concentrations with successive contactings with fresh soil is additional evidence that most of these elements are present as a colloidal dispersion and that they are readsorbed by the sediment in time, in agreement with the findings of Andelman and Rozzell<sup>13</sup> regarding coagulation of aged suspensions.

In summary, the following conclusions seem to be justified:

1. Plutonium and americium are dispersed from Pond B-1 sediment into Pond B-1 water in increasing amounts at pH 9 and above,

**TABLE 8. Successive 20-Hour Extractions of Original Sediment With Fresh Pond Water**

<u>Extraction</u>	<u>pH9</u>		<u>pH11</u>	
	<u>Pu Conc., pCi/l</u>	<u>Am Conc., pCi/l</u>	<u>Pu Conc., pCi/l</u>	<u>Am Conc., pCi/l</u>
<b>First</b>	173(±38)	60(±13)	1115(±235)	378(±61)
<b>Second</b>	74(±45)	34(±8)	1253(±170)	473(±40)
<b>Third</b>	9(±3)	~4	399(±53)	145(±53)
<b>Fourth</b>	15(±5)	8(±4)	198(±149)	60(±40)

**TABLE 9. Successive 20-Hour Extractions of Fresh Sediment With Original Pond Water**

<u>Extraction</u>	<u>pH9</u>		<u>pH11</u>	
	<u>Pu Conc., pCi/l</u>	<u>Am Conc., pCi/l</u>	<u>Pu Conc., pCi/l</u>	<u>Am Conc., pCi/l</u>
First	173(±38)	60(±13)	1115(±235)	378(±61)
Second	177(±12)	42(±10)	1695(±310)	445(±50)
Third	86(±30)	20(±10)	890(±170)	179(±27)
Fourth	34(±16)	15(±5)	393(±124)	51(±31)

and under certain conditions the concentrations of both elements can approach or even exceed Maximum Permissible Concentrations. (1667 pCi Pu/l, 1330 pCi Am/l)

2. The plutonium is dispersed as a colloidal sol, the average size of the colloid decreasing with increasing pH.
3. Americium is also present in colloidal form, and its size also decreases with increasing pH, but in a less regular manner.
4. Only a small percentage of the plutonium and americium in the sediment is present in a dispersable form.
5. Pond water has a relatively low saturation value for both elements, and at concentrations well below saturation, the colloid is gradually readsorbed by the sediment.
6. Because of items (4) and (5), migration of plutonium and americium down the pond outlet stream would be slow, but could cause problems over an extended period of time.
7. Because of item (4) it would probably not be feasible to remove plutonium and americium from Pond B-1 sediment by repeated leaching at high pH.
8. Although these results are specific for Pond B-1, they are probably applicable in varying degrees to other aquatic systems that are comparable in terms of history, chemistry, and biological conditions.

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ENVIRONMENTAL SCIENCES AND WASTE CONTROL  
SERVICE REPORT

Report No:

EC-317-74-127

Title:

SURVEY OF RESERVOIR SEDIMENTS.

Work Requested By: M. A. Thompson

Date Requested: June 1974

Work Performed By: K. K. Kunert and G. J. Werkema

Manhours: 80

Date Work Started: November 1973

Date Work Completed August 1974

Date This Report: August 23, 1974

Report Written By: K. K. Kunert and G. J. Werkema

REVIEWED FOR

FEB 7 1975

Patentable Material  
Time found  
by H. H. H. H.

Distribution

R. B. Gunning  
EMF (Record)

Thru: M. A. Thompson  
Environmental Master File

KWIC Index

Coring  
Cesium  
Plutonium  
Water-Sediments  
Sampling  
Environment

Reviewed for Classification/UCNI/OUO  
By: Janet Nesheim, Derivative Classifier  
DOE, EMCBC  
Date: 10-28-08 JAN  
Confirmed Unclassified, Not UCNI/Not OUO

~~INTERNAL REPORT~~

~~NOT CLEARED FOR PUBLICATION~~

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## INTRODUCTION

Core and dredge samples were collected from the bottoms of Great Western Reservoir and Standley Lake during October 1973. The sampling program was conducted jointly by the United States Environmental Protection Agency (USEPA) and the Colorado Department of Health<sup>1</sup> to assess selected radionuclide concentrations in the reservoir bottoms.

Core samples were collected in 1-inch diameter pipes which were sectioned in 1-inch horizons. Sediment (dredge) samples were collected using a Petersen dredge. Sample locations are shown in Figures 1 and 7.

The USEPA requested that Rocky Flats Plant participate in the analysis of approximately one-third of the core samples. The Rocky Flats portion of the samples were analyzed at Battelle-Pacific Northwest Laboratory and Lawrence Livermore Laboratory. The remaining samples were analyzed by the USEPA at the National Environmental Research Center-Las Vegas. The most complete data were obtained for plutonium and cesium-137.

Analytical data were exchanged with USEPA in June 1974. The following discussion is based on a graphical synthesis of the data, including horizontal dispersions at several depths, and vertical profiles at some stations.

## DISCUSSION

Tables I and II show the concentrations of Pu 239-240 and Pu 238 in the dredge samples and the core samples and the Cs 137 concentrations in the dredge samples in picocuries per gram (pCi/g). From these data contour drawings (Figures 2-6 and 7-9) were made of the Pu 239-240 and Cs 137 concentrations in the dredge samples and the Pu 239-240 concentrations for different depths of the core samples. Graphs were plotted of the concentration versus depth for the Pu 239-240 core samples (Figures 10-18).

The following conclusions are based on these graphical distributions and tables.

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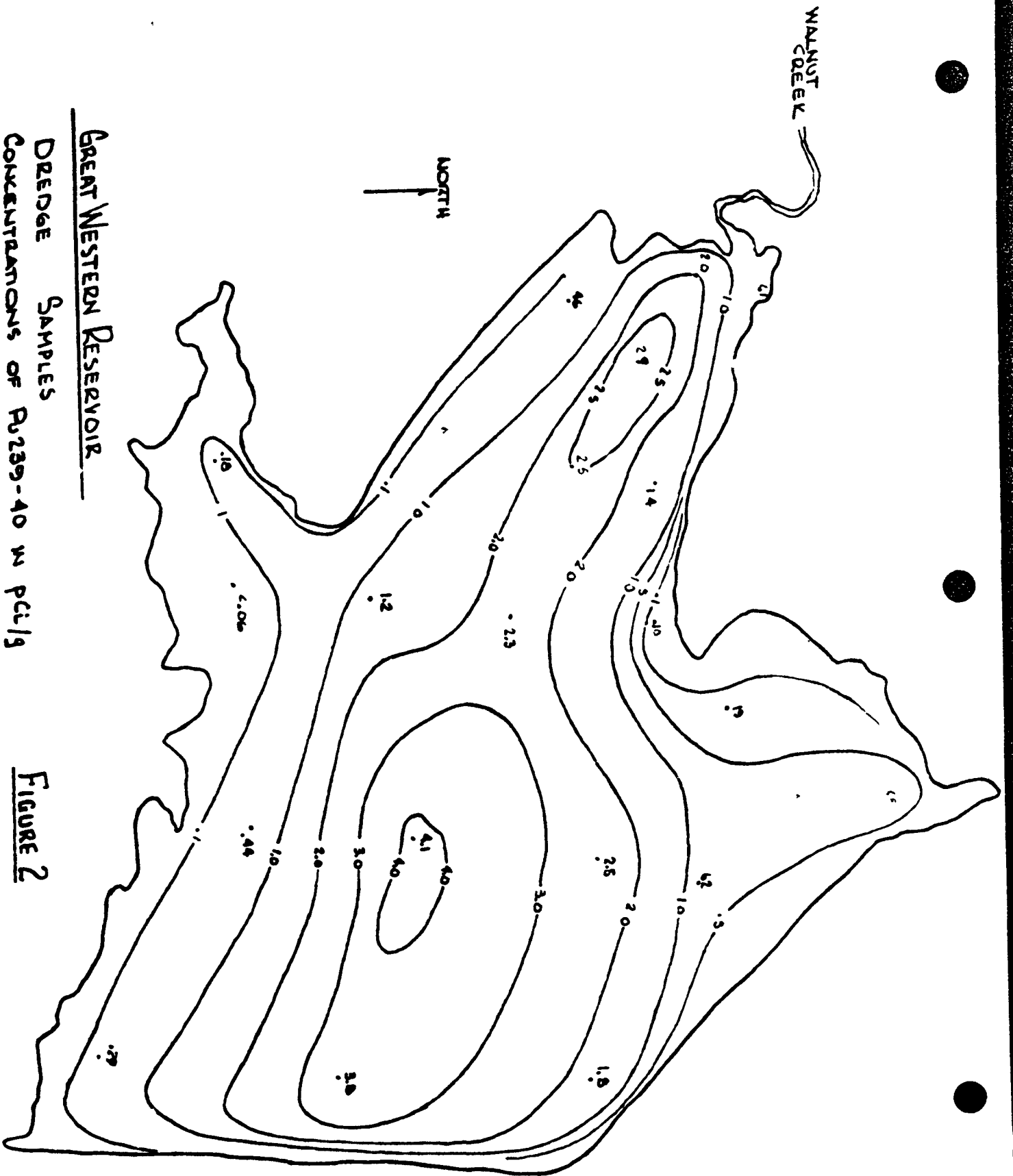
A. Great Western Reservoir

1. Highest concentrations of Pu 239-240 are found in the low areas of the reservoir and along the dam (Figures 2, 3, and 4).
2. Cs 137 concentrations are highest in the low area by the dam (Figure 6).
3. Core samples were not analyzed at every depth, so plots of concentration versus depth are not complete. In most cases the highest concentrations of Pu 239-240 were in the 0-1" level. At four sample locations the 1-2" level was higher than the 0-1" level. These locations are 1, 5, 8 and 20 (Figures 10, 12, 13, and 17).
4. Dredge samples 1, 12, and 21 (Table I) show a difference between the EPA's results and Rocky Flats' results, the EPA's being lower. In two cases, points 14 and 19, EPA and Rocky Flats results are close.

B. Standley Lake

1. Highest Pu 239-240 concentrations are found by the dam and by the island (Figure 8). These concentrations are lower than those for Great Western (0.1 for Standley and 4.1 for Great Western).
2. The Cs 137 concentrations in Standley Lake are about the same as in Great Western Reservoir (2.4 for Standley and 2.1 for Great Western). Cs 137 is also concentrated at the dam and around the island (Figure 9).
3. Sample points 2, 5, 7, 10, 15 and 17 show differences between EPA and Rocky Flats results, Rocky Flats results being higher (Table II).

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GREAT WESTERN RESERVOIR

DREDGE SAMPLES

CONCENTRATIONS OF  $Pu^{239-40}$  IN  $pCi/g$

FIGURE 2

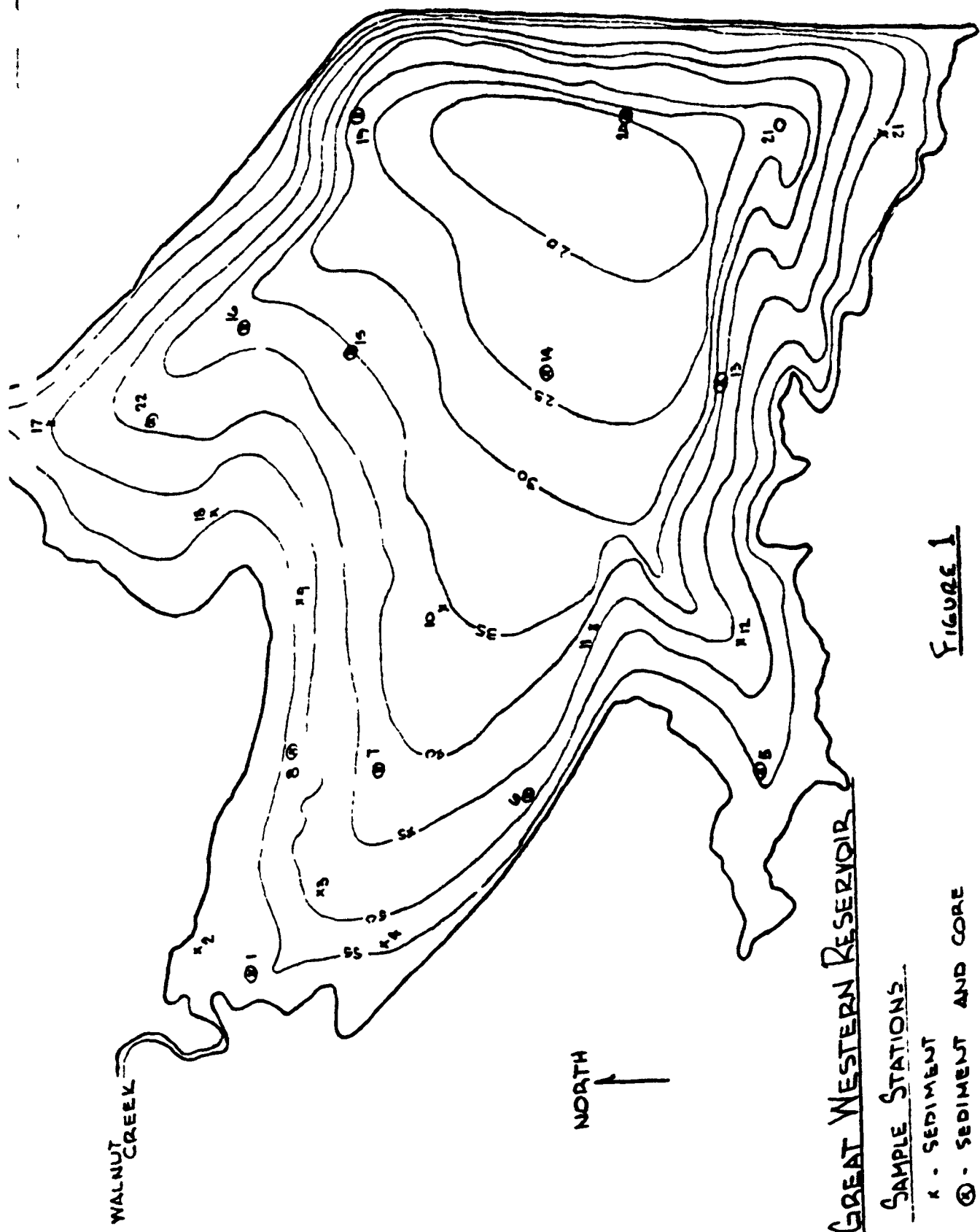


FIGURE 1

GREAT WESTERN RESERVOIR

SAMPLE STATIONS

- x - SEDIMENT
- O - SEDIMENT AND CORE